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NON-CRYSTALLINE COMPACT PACKINGS OF HARD SPHERES OF TWO SIZES: BIPYRAMIDS AND THE GEOMETRY OF COMMON NEIGHBOURS (PREPRINT)

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Metals Branch
Metals, Ceramics and NDE Division

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14. ABSTRACT

Insight into the efficient filling of space in systems of binary spheres is explored using bipyramids consisting of $3 \leq n \leq 8$ tetrahedra sharing a common pair of spheres. Compact packings are sought in bipyramids consisting of larger hard spheres of unit radius and smaller hard spheres of radius $0.001 \leq R \leq 1$. Seventy-seven distinct compact bipyramids are found. The number of distinct compact bipyramids increases with the number n of constituent tetrahedra. No compact bipyramids are found for $R \geq 0.9473$ and for $0.8493 \geq R \geq 0.7434$. A topological instability eliminates compact packings for $R \leq 0.1547$. Pentagonal bipyramids cover a larger range in R than any other compact bipyramids studied.

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Non-crystalline compact packings of hard spheres of two sizes: bipyramids and the geometry of common neighbours

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Abstract

Insight into the efficient filling of space in systems of binary spheres is explored using bipyramids consisting of $3 \leq n \leq 8$ tetrahedra sharing a common pair of spheres. Compact packings are sought in bipyramids consisting of larger hard spheres of unit radius and smaller hard spheres of radius $0.001 \leq R \leq 1$. Seventy-seven distinct compact bipyramids are found. The number of distinct compact bipyramids increases with the number n of constituent tetrahedra. No compact bipyramids are found for $R \geq 0.9473$ and for $0.8493 \geq R \geq 0.7434$. A topological instability eliminates compact packings for $R \leq 0.1547$. Pentagonal bipyramids cover a larger range in R than any other compact bipyramids studied.

1. Introduction

Efficient packing represents the most ubiquitous of ordering principles. While familiarity may have rendered the result commonplace, it is remarkable that the condition of density maximisation alone is sufficient to generate the face centered and hexagonal close packed crystals for a collection of identical hard spheres. The nature of the relationship between density maximization and the stability of *aperiodic* structures when the steric constraints become more complex is an open question which finds a sharp focus in considering the stability of metallic glasses with respect to crystallization. Here steric complexity is provided by the presence of spherical particles of different size. In this paper we propose that bipyramids represent useful local structural elements in amorphous packings of binary mixtures of hard spheres. We report on a survey of the sphere size ratios associated with all possible compact packings of 3-fold to 8-fold bipyramids.

Thinking about maximising the packing density of spheres of different size when periodic configurations are excluded, or at least not explicitly invoked, represents a major challenge from the outset. How is the problem to be posed? The most popular answer to this question has been to use model building – originally real models and now computer models – in which the optimization is carried out within the constraint of some modelling protocol. Pioneering this approach, Bernal [1] introduced the idea of 'random close packing' as being the maximum density of an amorphous collection of hard spheres of a single size. There have been a large number of studies of random close packing of hard spheres of one size [2,3] and two sizes [4,5]. Torquato et al [6] have criticised the reliance on the modelling protocol to *implicitly* define the amorphous constraint. These authors showed how their choice of modelling protocol imposed no such constraint and thus allowed a continuous path of sphere packings in terms of density from amorphous to fully crystalline.

The analysis of structure in computer modelling of disordered systems is often based on local organization of particles. Most of the literature makes use of one of three choices of local structural 'element': coordination polyhedra, common neighbours or tetrahedra. As these local measures form the basis of the geometrical approaches to amorphous packing described below, we shall briefly review the methods of structural analysis. Coordination polyhedra are defined using either Voronoi polyhedra [7] or their duals, the Delaunay tesselations [8]. The large variety of possible polyhedra tends to result in identifying a rather broad distribution of local structures in a liquid, many of which may only differ by one or two bonds. The common neighbour analysis was originally developed to resolve the geometrical origin of the first and second peaks in the pair distribution function of a liquid [9]. The idea is to consider all pairs of particles that share one or more neighbours. Honeycutt and Andersen [10] introduced a four integer notation to classify these common neighbours. The first integer is 1 or 2, depending on whether the root pair are in contact or not. The second integer indicates the number of neighbours common to the root pair. The third integer records the number of contacts among the common neighbours. A fourth integer is included that does not quantify some explicit topological feature but is used to distinguish between arrangements of the common neighbours that the first three numbers cannot differentiate. The close packed icosahedron consists of only a single unit, (1551), while the fcc crystal is made of the following common neighbour units: (2211), (2101), (1421) and (2441). (Note that in the case of the close packed crystal, the root pair are generally not in contact, as indicated by the first integer being two.)

Looking at ever smaller and, hence, more elementary, structural motifs we end up with tetrahedra. In 3D, the smallest number of spheres which can be identified as having a compact packing is four and that packing is a tetrahedron, an irregular one when spheres of different size are involved. Regular tetrahedra cannot be packed without leaving gaps [11]. Allowing for deviations away from regularity, however, Frank and Kasper [12] showed that it was possible to construct a range of polytetrahedral crystals. Bernal [1] estimated that the random close packed hard spheres were comprised of 86% tetrahedra, and a number of groups [11,13,14] have sought to link these facts by describing the structure of amorphous alloys in terms of disclination networks based on the Frank-Kasper analysis. Medvedev and coworkers [15] have analysed amorphous packings of spheres in terms of the network of tetrahedra connected through shared faces with other tetrahedra (and fragments of octahedra).

The considerations of local structure in amorphous packings described in the preceding paragraph suggest that, instead of statistically modelling amorphous packings of thousands of particles, it might be useful to determine the optimal packing geometries of the small number of particles involved in local structure. In this approach one explicitly solves for the densest packing of various local arrangements of particles. Just as a crystal structure can be resolved into the structure of a unit cell and the rules by which the unit cells are packed, so might aperiodic structures be resolved into some finite family of locally preferred structures [16] and the rules by which they can be assembled with one another to occupy space. This approach neglects the role of the particles that lie outside the local group.

Since the packing of spheres in a tetrahedron is straight forward, the choice of locally preferred structures have tended to fall into two groups. The first, with the most extensive literature, is the nearest neighbour coordination polyhedra. Frank adopted this approach in championing the role of icosahedral coordination [17]. Hoare and Pal [18] considered the close packing of spheres in clusters that extended well beyond the nearest neighbour coordination shell. Spheres of two sizes have been included to only a limited extent in studies of efficiently-packed clusters consisting of equal-sized spheres surrounding a central sphere of different size [19].

One problem with using polyhedra to resolve amorphous structures is that the loss or gain of a single edge (i.e. a contact between a pair of particles) changes the polyhedron without, necessarily, changing the structure that is being analysed in any significant way. As the polyhedra become larger, so do the number of such variants. To avoid this problem, we suggest looking at n-bipyramids, consisting of a pair of contacting spheres and their n common neighbours [10], as representing the smallest non-trivial packing element where frustration becomes important. The n-bipyramid consists of two axial sites and n equatorial sites, giving n tetrahedra that share the common axis and n dihedral angles common to the axial sites. An n-bipyramid is compact when each equatorial sphere contacts its two equatorial neighbours and the common neighbour pair. In the notation of Honeycutt and Andersen [10], these are common neighbour pairs of the type (1nn1). The n dihedral angles sum to 2π radians in these efficiently packed bipyramids, analogous to the compact packing in binary systems of 2D discs, where the planar triangles formed by the discs surrounding a common disc sum to 2π radians [20]. If there are just three common neighbours, we have a trigonal bipyramid; four common neighbours, an octahedron; five common neighbours, a pentagonal bipyramid, and so on. A number of papers [1,21] note that the pentagonal bipyramid occurs frequently in random close packed hard spheres. For simplicity, we consider binary bipyramids with larger hard spheres L of unit radius and smaller hard spheres S of radius $0.001 \leq R \leq 1$. Compact bipyramids are reported here, along with topological characteristics including the relative sphere sizes, relative concentration and specific bipyramid configurations. We specifically address the packing within bipyramids, but do not address the packing between bipyramids.

2. Approach

Establishing close packing in a particular *n*-bipyramid, it is sufficient to show that the *n* dihedral angles about the axial pair of particles sum to 2π. The dihedral angles are explicit functions of the S and L sphere radii. Consider the tetrahedron OABC in Figure 1, where O, A, B and C are centres of S or L spheres and OA is the tetrahedron edge about which the dihedral angle, *D*, is measured. The dihedral angle can be expressed in terms of the three planar angles: BOC (α), AOC (β) and AOB (γ) as follows,

$$D = \cos^{-1}\left\{\frac{\cos\alpha - (\cos\beta)(\cos\gamma)}{(\sin\beta)(\sin\gamma)}\right\} \qquad 1$$

The planar angles can, in turn, be expressed in terms of the separation between sphere centres when in contact as follows,

$$\cos\alpha = \left\{\frac{|OB|^2 + |OC|^2 - |BC|^2}{2|OB||OC|}\right\} \qquad \text{2a}$$

$$\cos\beta = \left\{\frac{|OA|^2 + |OC|^2 - |AC|^2}{2|OA||OC|}\right\} \qquad \text{2b}$$

$$\cos\gamma = \left\{ \frac{|OA|^2 + |OB|^2 - |AB|^2}{2|OA||OB|} \right\} \qquad \text{2c}$$

where the edge length $|OA|$ is the sum of sphere radii that occupy O and A. There are three possible edge lengths (2R, 1+R and 2) and five planar angle cosines (½, $R/(1+R)$, $1-2/(1+R)^2$, $1/(1+R)$ and $1-[2R/(1+R)]^2$). Bipyramids are specified by the two spheres that define the common axis and by the sequence of n equatorial spheres. For each axis pair, the three possible combinations of two equatorial spheres give three possible dihedral angles, D_{SS}, D_{SL} and D_{LL}. The packing in bipyramids of a given axis pair can thus be described by a linear combination of these angles

$$iD_{SS} + jD_{SL} + kD_{LL} \qquad 3$$

where i, j and k represent the number of dihedral angles with SS, SL and LL equatorial pairs, respectively. The distinct equatorial sequences are listed in Table I, along with the i, j, k values for each distinct sequence. There are three axis pairs for each equatorial sequence, and the resulting number of possible n-bipyramids is shown in Table II. Several unique equatorial sequences have identical i, j, k indices when $n \geq 6$, and so give identical packing. The number of distinct i, j, k packings is also shown in Table II.

We search for compact solutions in each possible n-bipyramid $3 \leq n \leq 8$ over the smaller radius R such that $0.001 \leq R \leq 1$. Compact bipyramids are specified by the axis pair, by the distinct sequence of equatorial spheres and by the S sphere radius, R.

3. Results

In Tables III-VIII we present our results – the specific bipyramids, the value of R at which they are compact and the fraction of S spheres, F_s. Nearly all of the equatorial sequences in Table I give compact packings for at least one of the three axial pairs. Those that do not are listed in italics. Compact bipyramids exist only for SS or SL axis pairs when $n \leq 5$ (Tables III-V), and only for the LL axis pair when $n \geq 6$ (Tables VI-VIII).

The R values for which compact bipyramids are obtained are plotted in Figure 2 for each n value studied. Compact bipyramids with $R > 2/3$ exist only for pentagonal bipyramids. There are no compact packings above $R = 0.9473$. This maximum value is achieved by the pentagonal bipyramid consisting of 5 L equatorial spheres with an SS axis. For this pentagonal bipyramid, R decreases to as low as 0.7434 and F_s increases as S spheres are added to the equatorial ring (Figure 3). The same trend is repeated for pentagonal bipyramids with the SL axis pair, starting at $R = 0.9022$ for the bipyramid with 5 L equatorial spheres, and continuing to a lower bound of $R = 0.2236$ for 2 L and 3 S equatorial spheres. The pentagonal bipyramids cover the largest range of R for any of the compact n-bipyramids studied.

Compact bipyramids with $R \leq 2/3$ are dominated by $n = 6, 7, 8$. These bipyramids cover a nearly continuous span of R from 0.6667 to 0.1553, the lowest R obtained in this study. Far fewer compact bipyramids are found with $n = 3$ or 4, which sparsely cover the range $0.5000 \geq R \geq 0.1667$. As shown in Figure 3, a decrease in R is achieved by an increase in F_s when $n \leq 5$, and is accomplished by a decrease in F_s when $n \geq 6$.

The gaps are real. In addition to the upper limit of R = 0.9473, no compact packings are produced over the range 0.8493 ≥ R ≥ 0.7434. The abrupt absence of compact packings below R ≤ 0.1553 arises from a fundamental instability in tetrahedra with 3L and 1S spheres. The S sphere just fills the interstice between the 3L spheres when $R = \left(2/\sqrt{3}\right) - 1 = 0.1547...$, forming a planar trigonal array rather than a tetrahedron. This instability affects bipyramids with the SL axis pair when $k \neq 0$ and the LL axis pair when $j \neq 0$, eliminating over half the SL bipyramids and almost all the LL bipyramids for $R < \left(2/\sqrt{3}\right) - 1$. When $R < \left(2/\sqrt{3}\right) - 1$, bipyramid packings approach the compact state as $R \to 0$ for the SS axis pair with the SLL and SLSL equatorial sequences and for the SL axis pair with the SLSL and SSSSL equatorial sequences.

These bipyramids can be used to construct clusters. In the simplest way, two identical bipyramids are combined by sharing a common axis sphere which then forms the central sphere of the cluster. In this way, two bipyramids with the SL/LLLLL axis pair/equatorial spheres configuration that share the common S sphere are used to form an icosahedron with a central S surrounded by 12L. Clusters produced in this way need not be efficiently packed, as illustrated by combining the same two SL/LLLLL bipyramids into a cluster that shares the common L of the axis pair. Non-identical bipyramids can also be combined, but packing frustration from such combinations is likely to be common.

The SS/SSLL and SL/SSLL bipyramids are both compact at R = 0.1716. Each of these two distinct bipyramids is a portion of a cluster comprised of an inner tetrahedron of 4S enclosed by a tetrahedron of 4L. Each L nestles in the centre of each of the 4 faces formed by the smaller tetrahedron, and the 4L spheres just contact each other.

A single plane passes through all *n* equatorial sphere centres for bipyramids with the SS and LL axis pairs, and this plane bisects the line defined by the SS and LL axis pair centres. For the SL axis pair, the equatorial plane is normal to the axis only when all of the equatorial spheres are of the same type. However, this plane no longer bisects the axis, and if S is sufficiently small, it need not intersect the SL axis at all. In general, a single plane does not pass through all *n* equatorial centres in bipyramids with an SL axis pair and mixed equatorial spheres.

4. Discussion

In 1987 Honeycutt and Andersen [10] reported on the common neighbour distribution in a supercooled binary mixture of Lennard-Jones particles with a size ratio of 0.8. They observed that the number of contact pairs with five common neighbours (i.e. 1551 or pentagonal bipyramids) increased significantly on cooling. At the lowest temperature reported, 61% of the particles were involved in either a 1551 or a 2331 common neighbour pair (the two common neighbour environments found in an icosahedron). The significance of the pentagonal bipyramids lends support to our proposition that the close packing of bipyramids represent useful description of the close packing in extended amorphous phase.

Honeycutt and Andersen went on to identify the pentagonal bipyramids as icosahedral environments. Was this justified? Consider, for example, the bipyramid with S particles in both axial positions and the equatorial sequence SSLSL. This bipyramid is close-packed when

$R = 0.8678$. Decorating part of the icosahedral net with this bipyramid we see that each large particle sits on an axial position of a new bipyramid with a small particle in the other axial position and an equatorial sequence SSSxx. The last two equatorial positions are unspecified but, looking through the list of possible close packed pentagonal bipyramids, we see that the only bipyramid that meets these conditions is one with the equatorial sequence SSSLL. This new bipyramid is close-packed when $R_2 = 0.4202$, a size ratio significantly smaller than that of the original bipyramid so we can conclude that the bipyramid SS/SSLSL cannot be involved in a close-packed polyhedra on an icosahedral net. Based on similar reasoning we conclude that out of the 13 close-packed pentagonal bipyramids, only three: SS/LLLLL, SL/SLLLL and SL/LLLLL, could provide the basis for dense packing of a polyhedron with icosahedral topology, and only the last can actually produce a close packed icosahedron.

The preceding argument demonstrates that the presence of pentagonal bipyramids in an amorphous packing does not imply the presence of icosahedra. This is, of course, not the same as proving that icosahedra are absent in such an amorphous state, just that, if present, they include elements other than the compact bipyramids treated here. The Laves crystal $MgZn_2$, for example, represents a reasonable AB_2 packing (with a packing fraction close to that of fcc) and contains icosahedral coordination polyhedra about the smaller (B) particle in spite of the inability of the pentagonal bipyramids to form such a polyhedron at this size ratio. In fact, the same model studied by Honeycutt and Andersen has recently [22] been shown to freeze into the $MgZn_2$ structure and, in fact, to have a substantial amount of icosahedral coordination in the supercooled liquid. These icosahedra, far from stabilizing the liquid from freezing as envisioned by Frank, are actually the precursors of the crystal phase.

We propose here that compact bipyramids are structural elements that may be important in maximising the packing efficiency in disordered systems of binary spheres– a problem of significant relevance. Decreasing packing efficiency within a given bipyramid is expected for increasing deviation from discrete values of *R* that give compact bipyramids. Although the number of compact bipyramids is rather small, giving a small set of discrete *R* values, these values are nevertheless fairly evenly distributed over the bounding interval of $0.1547 \leq R \leq 0.9473$ (Figure 2). Thus, bipyramids can generally be produced for any *R* value that is not far from one that gives compact packing. The gap over the interval $0.7434 \leq R \leq 0.8493$ is notable, in that it is significantly larger than any other interval that excludes compact bipyramids. This gap represents a range in *R* over which bipyramids cannot be nearly efficiently packed, and this may have relevance in the efficient packing in disordered systems of binary spheres.

5. Conclusions

Insight into the efficient filling of space in systems of unequal spheres is explored using bipyramids constructed of 3 to 8 tetrahedra that share a common pair of spheres. Two sphere sizes are used– a larger sphere L with a fixed radius of unity and a smaller sphere S with radius $0.001 \leq R \leq 1$. Seventy-seven distinct compact bipyramids are identified. Two distinct compact bipyramids are found for $n = 3$, 6 for $n = 4$, 13 for $n = 5$, 12 for $n = 6$, 17 for $n = 7$ and 27 for $n = 8$. The largest *R* that produces compact packing is 0.9473. No compact bipyramids are found over the interval $0.8493 \geq R \geq 0.7434$. A topological instability eliminates compact packings for

$R \leq 0.1547$. Compact pentagonal bipyramids are found over a larger range in R than any other bipyramids studied.

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FIGURE CAPTIONS

Figure 1. The dihedral AOBC of a bipyramid cluster

Figure 2. Size ratios for which compact packings exist for various bipyramids. Bipyramids are identified by both the number of particles in the equatorial positions and the type of particles in the two axial sites: SS (●), SL (▽) and LL (◆). The size ratios for all compact bipyramids described here are shown at the bottom of the figure (▲).

Figure 3. The fraction of small spheres F_s as a function of R for the type of particles in the two axial sites: SS (●), SL (▽) and LL (◆).

n = 3		**n = 4**		**n = 5**		**n = 6**		**n = 7**		**n = 8**	
SSS	*3,0,0*	*SSSS*	*4,0,0*	*SSSSS*	*5,0,0*	SSSSSS	6,0,0	SSSSSSS	7,0,0	SSSSSSSS	8,0,0
SSL	*1,2,0*	*SSSL*	*2,2,0*	SSSSL	3,2,0	SSSSSL	4,2,0	SSSSSSL	5,2,0	SSSSSSSL	6,2,0
SLL	*0,2,1*	SSLL	1,2,1	SSSLL	2,2,1	SSSSLL	3,2,1	SSSSSLL	4,2,1	SSSSSSLL	5,2,1
LLL	0,0,3	SLSL	0,4,0	SSLSL	1,4,0	SSSLSL	2,4,0	SSSSLSL	3,4,0	SSSSSLSL	4,4,0
		SLLL	0,2,2	SSLLL	1,2,2	SSLSSL	2,4,0	SSSLSSL	3,4,0	SSSSLSSL	4,4,0
		LLLL	*0,0,4*	SLSLL	0,4,1	SSSLLL	2,2,2	SSSSLLL	3,2,2	SSSLSSSL	4,4,0
				SLLLL	0,2,3	SSLSLL	1,4,1	SSSLSLL	2,4,1	SSSSSLLL	4,2,2
				LLLLL	0,0,5	SLSLSL	0,6,0	SSLSSLL	2,4,1	SSSSLSLL	3,4,1
						SSLLLL	1,2,3	SSLSLSL	1,6,0	SSSLSSLL	3,4,1
						SLSLLL	0,4,2	SSSLLLL	2,2,3	SSSLSLSL	2,6,0
						SLLSLL	0,4,2	SSLSLLL	1,4,2	SSLSSLSL	2,6,0
						SLLLLL	0,2,4	SSLLSLL	1,4,2	SSSSLLLL	3,2,3
						LLLLLL	*0,0,6*	SLSLSLL	0,6,1	SSSLSLLL	2,4,2
								SSLLLLL	1,2,4	SSLSSLLL	2,4,2
								SLSLLLL	0,4,3	SSSLLSLL	2,4,2
								SLLSLLL	0,4,3	SSLLSSLL	2,4,2
								SLLLLLL	0,2,5	SSLSLSLL	1,6,1
								LLLLLLL	*0,0,7*	SSLSLLSL	1,6,1
										SLSLSLSL	0,8,0
										SSSLLLLL	2,2,4
										SSLSLLLL	1,4,3
										SSLLSLLL	1,4,3
										SLSLSLLL	0,6,2
										SLSLLSLL	0,6,2
										SSLLLLLL	*1,2,5*
										SLSLLLLL	0,4,4
										SLLSLLLL	0,4,4
										SLLLSLLL	0,4,4
										SLLLLLLL	*0,2,6*
										LLLLLLLL	*0,0,8*

Table I. Equatorial sphere sequences and corresponding i,j,k values in bipyramids

n	Number of distinct bipyramids	Number of distinct *i, j, k* packings	Number of compact bipyramids in $0.001 \le R \le 1$
3	12	12	2
4	18	18	6
5	24	24	13
6	39	33	12
7	54	42	17
8	90	54	27

Table II. Number of distinct bipyramids, distinct packings and compact bipyramids

Axial spheres	Equatorial sphere sequence	i, j, k	F_s	R
SS	LLL	0, 0, 3	0.4	0.1667
SL	LLL	0, 0, 3	0.2	0.2247

Table III. Compact trigonal bipyramids (n = 3)

Axial spheres	Equatorial sphere sequence	i, j, k	F_s	R
SS	SSLL	1, 2, 1	0.667	0.1716
SS	SLLL	0, 2, 2	0.5	0.3625
SS	LLLL	0, 0, 4	0.333	0.5000
SL	SSLL	1, 2, 1	0.5	0.1716
SL	SLLL	0, 2, 2	0.333	0.2808
SL	LLLL	0, 0, 4	0.167	0.4142

Table IV. Compact quadrilateral bipyramids (n = 4)

Axial spheres	Equatorial sphere sequence	i, j, k	F_s	R
SS	SSSSL	3, 2, 0	0.857	0.7434
SS	SSSLL	2, 2, 1	0.714	0.8710
SS	SSLSL	1, 4, 0	0.714	0.8678
SS	SSLLL	1, 2, 2	0.571	0.9129
SS	SLSLL	0, 4, 1	0.571	0.9119
SS	SLLLL	0, 2, 3	0.429	0.9342
SS	LLLLL	0, 0, 5	0.286	0.9473
SL	SSSLL	2, 2, 1	0.571	0.4202
SL	SSLSL	1, 4, 0	0.571	0.2236
SL	SSLLL	1, 2, 2	0.429	0.7206
SL	SLSLL	0, 4, 1	0.429	0.6902
SL	SLLLL	0, 2, 3	0.286	0.8493
SL	LLLLL	0, 0, 5	0.143	0.9022

Table V. Compact pentagonal bipyramids ($n = 5$)

Axial spheres	Equatorial sphere sequence	i, j, k	F_s	R
LL	SSSSSS	6, 0, 0	0.75	0.6667
LL	SSSSSL	4, 2, 0	0.625	0.6247
LL	SSSSLL	3, 2, 1	0.5	0.5591
LL	SSSLSL	2, 4, 0	0.5	0.5774
LL	SSLSSL	2, 4, 0	0.5	0.5774
LL	SSSLLL	2, 2, 2	0.375	0.4716
LL	SSLSLL	1, 4, 1	0.375	0.5034
LL	SLSLSL	0, 6, 0	0.375	0.5275
LL	SSLLLL	1, 2, 3	0.25	0.3600
LL	SLSLLL	0, 4, 2	0.25	0.4142
LL	SLLSLL	0, 4, 2	0.25	0.4142
LL	SLLLLL	0, 2, 4	0.125	0.2454

Table VI. Compact hexagonal bipyramids ($n = 6$)

Axial spheres	**Equatorial sphere sequence**	**i, j, k**	**F_s**	**R**
LL	SSSSSSS	7, 0, 0	0.778	0.4638
LL	SSSSSSL	5, 2, 0	0.667	0.4300
LL	SSSSSLL	4, 2, 1	0.556	0.3684
LL	SSSSLSL	3, 4, 0	0.556	0.3996
LL	SSSLSSL	3, 4, 0	0.556	0.3996
LL	SSSSLLL	3, 2, 2	0.444	0.2999
LL	SSSLSLL	2, 4, 1	0.444	0.3427
LL	SSLSSLL	2, 4, 1	0.444	0.3427
LL	SSLSLSL	1, 6, 0	0.444	0.3739
LL	SSSLLLL	2, 2, 3	0.333	0.2318
LL	SSLSLLL	1, 4, 2	0.333	0.2856
LL	SSLLSLL	1, 4, 2	0.333	0.2856
LL	SLSLSLL	0, 6, 1	0.333	0.3235
LL	SSLLLLL	1, 2, 4	0.222	0.1787
LL	SLSLLLL	0, 4, 3	0.222	0.2345
LL	SLLSLLL	0, 4, 3	0.222	0.2345
LL	SLLLLLL	0, 2, 5	0.111	0.1553

Table VII. Compact heptagonal bipyramids (n = 7)

Axial spheres	Equatorial sphere sequence	i, j, k	F_s	R
LL	SSSSSSSS	8, 0, 0	0.8	0.3431
LL	SSSSSSSL	6, 2, 0	0.7	0.3246
LL	SSSSSSLL	5, 2, 1	0.6	0.2746
LL	SSSSSLSL	4, 4, 0	0.6	0.3103
LL	SSSSLSSL	4, 4, 0	0.6	0.3103
LL	SSSLSSSL	4, 4, 0	0.6	0.3103
LL	SSSSSLLL	4, 2, 2	0.5	0.2250
LL	SSSSLSLL	3, 4, 1	0.5	0.2679
LL	SSSLSSLL	3, 4, 1	0.5	0.2679
LL	SSSLSLSL	2, 6, 0	0.5	0.2993
LL	SSLSSLSL	2, 6, 0	0.5	0.2993
LL	SSSSLLLL	3, 2, 3	0.4	0.1824
LL	SSSLSLLL	2, 4, 2	0.4	0.2288
LL	SSSLLSLL	2, 4, 2	0.4	0.2288
LL	SSLSSLLL	2, 4, 2	0.4	0.2288
LL	SSLLSSLL	2, 4, 2	0.4	0.2288
LL	SSLSLSLL	1, 6, 1	0.4	0.2633
LL	SSLSLLSL	1, 6, 1	0.4	0.2633
LL	SLSLSLSL	0,8,0	0.4	0.2910
LL	SSSLLLLL	2, 2, 4	0.3	0.1573
LL	SSLSLLLL	1, 4, 3	0.3	0.1962
LL	SSLLSLLL	1, 4, 3	0.3	0.1962
LL	SLSLSLLL	0, 6, 2	0.3	0.3235
LL	SLSLLSLL	0, 6, 2	0.3	0.3235
LL	SLSLLLLL	0, 4, 4	0.2	0.1726
LL	SLLSLLLL	0, 4, 4	0.2	0.1726
LL	SLLLSLLL	0, 4, 4	0.2	0.1726

Table VIII. Compact octagonal bipyramids ($n = 8$)

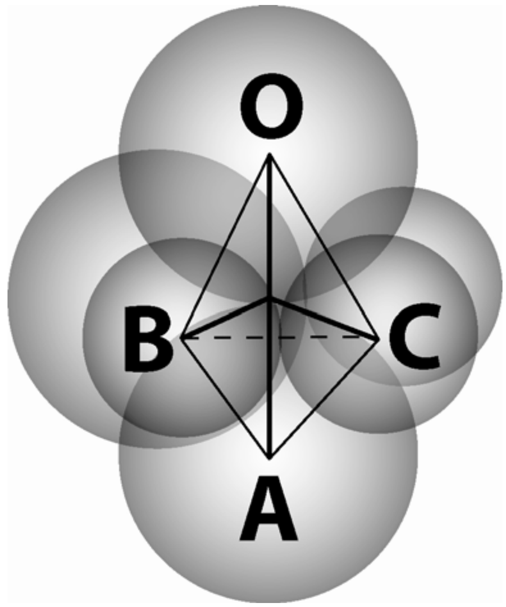

Figure 1. The dihedral AOBC of a bipyramid cluster

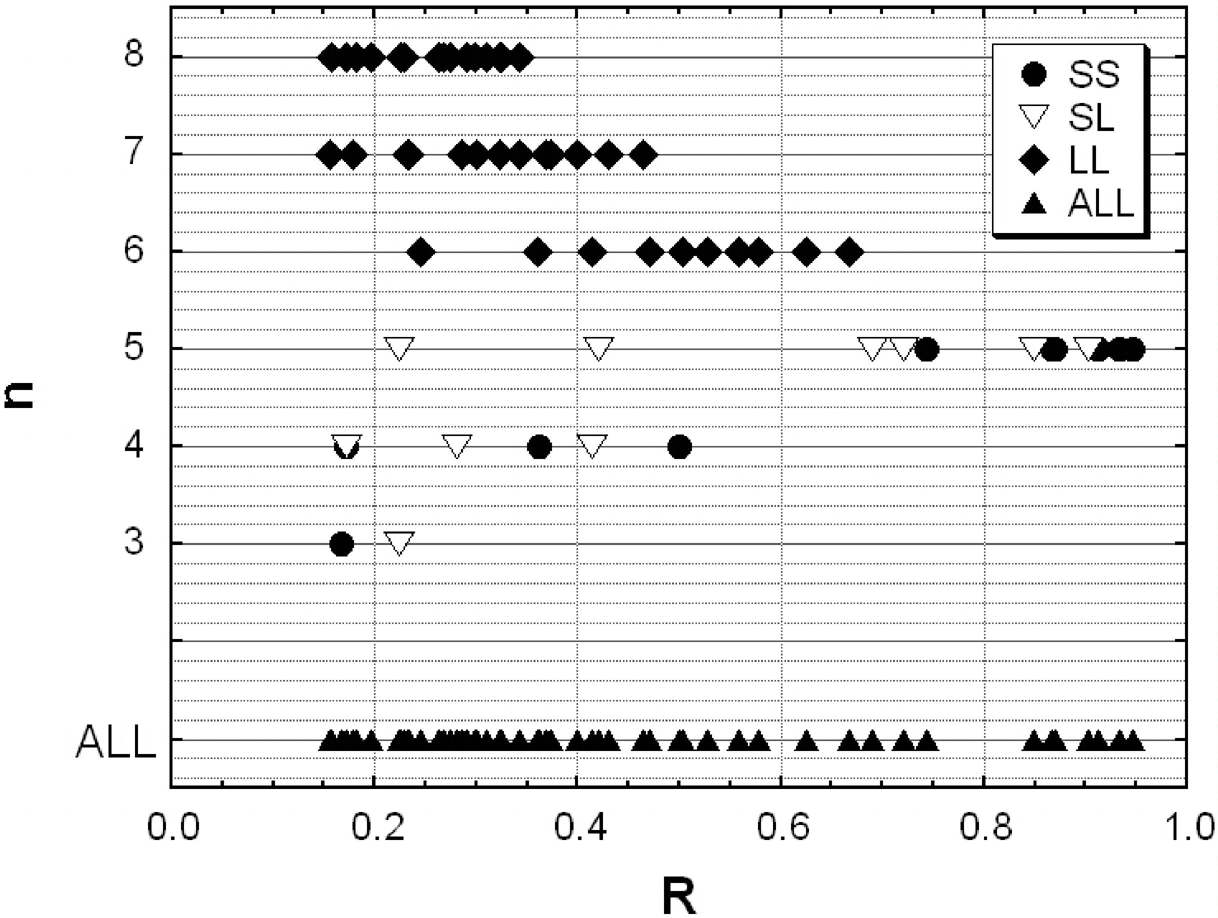

Figure 2. Size ratios for which compact packings exist for various bipyramids. Bipyramids are identified by both the number of particles in the equatorial positions and the type of particles in the two axial sites: SS (●), SL (▽) and LL (◆). The size ratios for all compact bipyramids described here are shown at the bottom of the figure (▲).

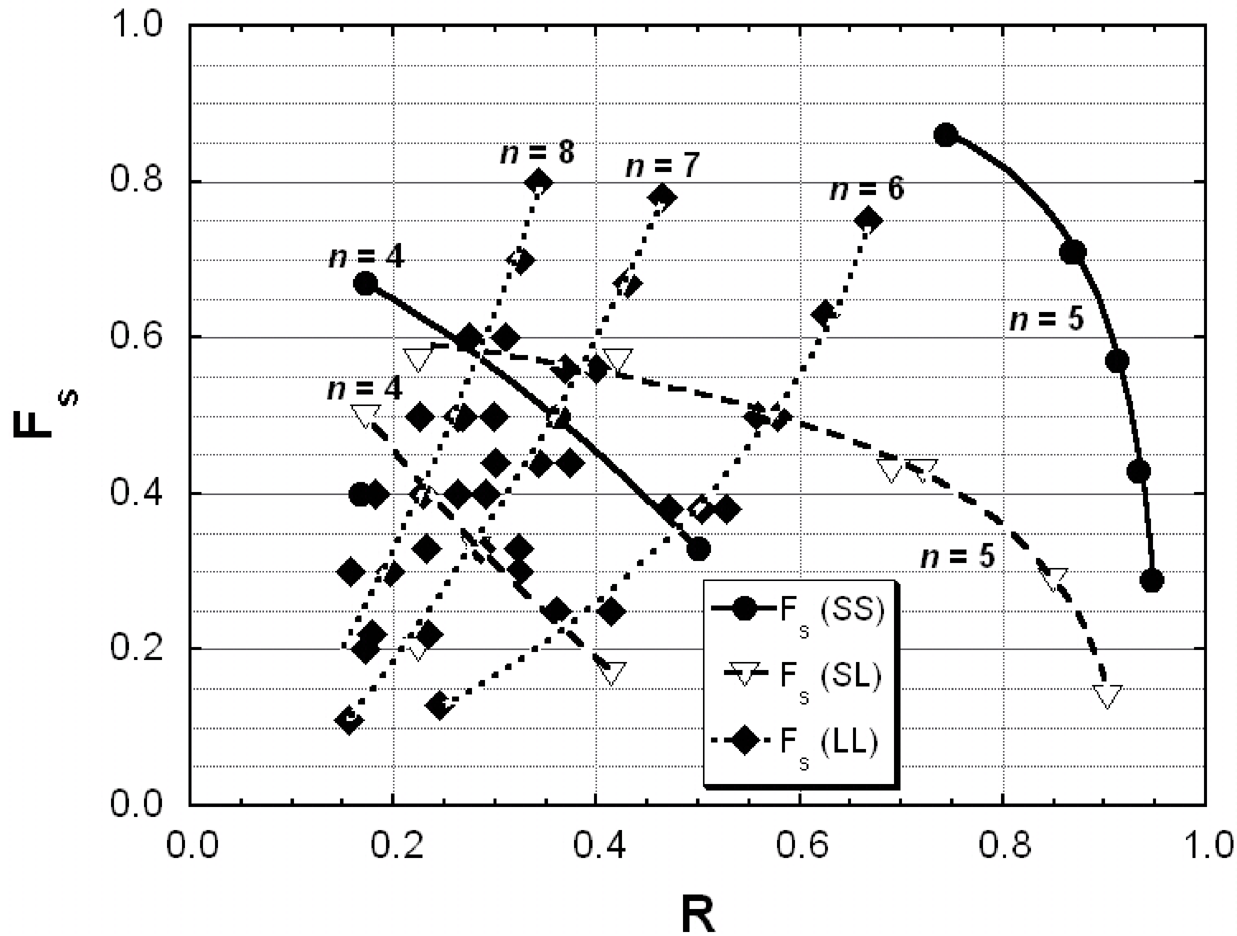

Figure 3. The fraction of small spheres F_s as a function of R for the type of particles in the two axial sites: SS (●), SL (▽) and LL (◆).